Powerful CO_2 methanation system by electrically heating spiral-type structured catalyst

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Significance and Relevance

This study demonstrates an innovative CO_2 methanation system using a spiral- type structured catalyst, which is directly heated by an electric current. The constructed methanation system indicated a powerful and stable performance even under a severe condition. Infrared imaging confirmed a desirable temperature profile with uniform distribution along with the spiral shape. Notably, the optimization of the spiral shape such as a twisting angle degree significantly affected on the methanation performance. Long-term stability tests confirmed CO_2 conversion over 50 hours at 350 °C.

Introduction and Motivations

The CO_2 methanation reaction ($CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$) has gained attention as a key technology for Carbon Capture and Utilization (CCU), allowing the conversion of CO_2 into valuable methane. Our research group focuses on the industrial implementation of this reaction, particularly for flue gas, which typically contains oxygen that can degrade catalysts through oxidation. However, we have discovered that oxygen can enhance CO_2 methanation.[1,2] The presence of oxygen induces an auto-methanation (AM) process, where the combustion of hydrogen and oxygen generates internal heating in the reactor, driving the methanation reaction without external heating. This internal heating increases energy efficiency. A key innovation in this research is the use of spiral catalysts, which are commonly used to improve heat transfer in catalytic systems. These catalysts facilitate the efficient delivery of thermal energy to the reaction site through the swirling flow of the gas. The metallic helical base material of the catalyst enhances heat conduction, which significantly improves reaction efficiency.

In this study, we present a new method of internal heating called 'electrical heating' (*e*-reaction), where the catalyst itself is powered by electricity and generates reaction heat by Joule heating. The schematic diagram of methanation with electrical heating (*e*-methanation) system is shown in Figure 1. Spiral-type catalysts, typically made of relatively high-resistance Ni-Cr metals, are ideal for this purpose. When an electric current is applied, the catalyst forms a heated reaction field. This method allows a uniform temperature distribution within the reactor, achieving rapid heating and improving reaction efficiency, potentially at lower power input. Thus, in this study, the reaction characteristics of the e-methanation system were investigated, as well as the effect of twisting angle of the spiral catalyst. In addition, the durability of the system was evaluated.

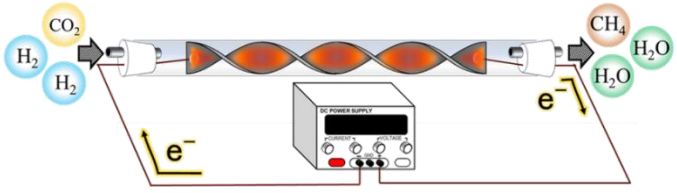

Figure 1 Schematic diagram of e-reaction system.

Materials and Methods

A wash-coat method was used to prepare the spiral-shaped catalysts. Ru/CeO_2 powder was obtained by impregnating and calcining the Ru component onto a CeO_2 substrate with a 10 wt% Ru loading. The powder catalyst was then suspended in distilled water, and a spiral substrate (11 mm wide × 180 mm long) twisted to 720° was immersed in the suspension and dried overnight. The spiral base material is a porous metal with continuous pores. This spiral base material was dipped and dried several times to obtain a spiral-shaped catalyst with a coating weight of 1.0 g·piece^{-1}.

The catalyst underwent hydrogen reduction at 200 °C for 60 min. The temperature was subsequently increased by applying 10 W of power in an N_2 atmosphere. When a constant temperature

was reached, the reaction gas was supplied. The power was gradually decreased when the reaction reached a steady state.

Results and Discussion

Figure 2 shows the CO_2 conversion and average temperature of the spiral catalyst when the power input, initially 10 W, was gradually reduced to 5 W. The average measured temperature at each position (expressed as the average temperature) is shown in the figure. At 10 W, the CO_2 conversion reached 80% with an average temperature of 297 °C. Subsequently, the power input was gradually reduced. Even at 5 W, the CO_2 conversion remained high at 75%, with an average temperature of 210 °C, approximately 90 °C lower than at 10 W.

Figure 3 shows the effect of varying the twist angle (0°–1080°) on methane conversion properties for plate and spiral catalysts. Higher twist angles led to increased CO_2 conversion, likely due to stronger swirl flow on the catalyst surface, improving methanation efficiency. Spiral heat exchangers are effective heat conductors, with their geometry promoting swirl flow, reducing boundary film resistance, and increasing the heat transfer coefficient. The induced turbulent gas flow thins the boundary layer, a major factor in increasing heat exchange efficiency. Therefore, higher twist angles increase methanation efficiency by lowering gas boundary films. The fact of no significant difference in reactivity with increasing angle is predicted to indicate that the swirling effect is sufficient at 360° of twist angle. It is expected that the effect of twist angle would be demonstrated under conditions where the effect of mass transfer is significant.

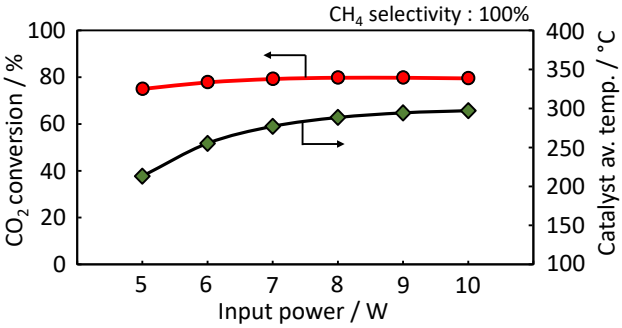

Figure 2 CO_2 conversion and average temperature of catalyst at each power input power.

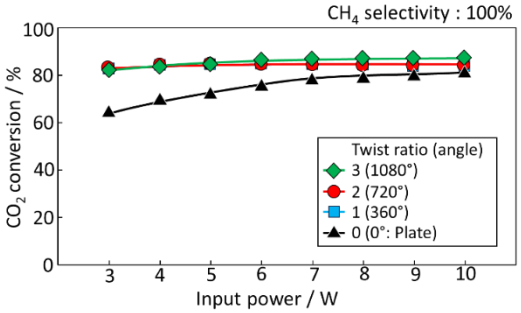

Figure 3 CO_2 conversion of spiral structured catalyst at an initial power input of 10 W.

Figure 4 shows the results of the e-methanation over an extended period, with power input fixed at 5 W and the reaction characteristics evaluated after 50 h. In the early stages of the reaction, a slight decrease in conversion was observed, but the performance remained almost constant. The average temperature of the reaction field was maintained at 350 °C. This consistent temperature, maintained by the effective current flow through the substrate and the swirling flow generated by the spiral shape of the catalyst bed, providing a sufficient downstream heating of the catalyst bed and high conversion characteristics.

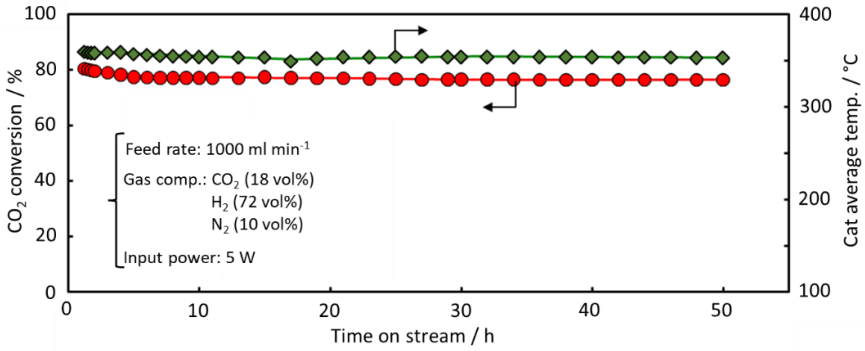

Figure 4 Durability performance for *e*-Methanation.

References

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